Investigation of Metastable Xenon grant: N66001-06-1-2041

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Abstract

In this investigation, we observe magnetic resonance signals of metastable ^{129}Xe sealed in a pyrex cell. We use external RF electrodes to generate the metastable Xe atoms. The metastable ^{129}Xe is optically pumped by a diode laser tuned to the optical resonance of the metastable state. The hyperfine resonance signal of metastable ^{129}Xe has been observed for the first time in a discharge cell, and a 20% polarization of metastable ^{129}Xe atoms has been measured. We also measured the linewidths of the Zeeman transitions and hyperfine transitions, which we compare with previous measurements of Zeeman resonances.

1 Introduction and optical spectroscopy

The electronic configuration of metastable Xe consists of tightly bound core electrons in the first 5 shells with an electron hole in $5P_{1/2}$ or $5P_{3/2}$ and a valence electron in the S orbital of the next shell $6S_{1/2}$. There are different possible couplings of angular momentum of two different electron holes and the valence electron. We denote the coupled state as $[5P_j, 6S_{1/2}]_J$. The four possible jj couplings are $[5P_{1/2}, 6S_{1/2}]_0$, $[5P_{1/2}, 6S_{1/2}]_1$, $[5P_{3/2}, 6S_{1/2}]_1$ and $[5P_{3/2}, 6S_{1/2}]_2$. In this investigation, we are interested in the lowest energy level $[5P_{3/2}, 6S_{1/2}]_2$ with electronic angular momentum $J = 2$, which is the metastable state. The lifetime of this energy level in our cells is not determined by spontaneous decay [1] but by the rate of collisions with other atoms or the cell wall, in which the valence electron releases energy and recombines with the hole. This state of ^{129}Xe exhibits some similarity with the ground state of an alkali metal. Metastable Xe can be optically pumped into a higher excited state by a near IR laser or lamp. The optical excitation is from the metastable state to an excited state in which the valence electron is in the $6P$ orbital. In the excited state, the total angular momentum is the coupling of valence electron spin and L', where L' is an intermediate coupling of valence electron orbital angular momentum and the total angular momentum of the hole in $5P$. We denote the coupled state $[L']_J$ when the valence electron is pumped into $6P$ orbital. The wavelengths of the optical excitation from metastable state to $[3/2]_2$ and $[5/2]_3$ are 823 nm (Fig. 1) and 882 nm (Fig. 2) respectively. The abundance of ^{129}Xe in the sample is 86%, and the rest of the isotopes are even isotopes except for ^{131}Xe. Among all the xenon isotopes in the cell, only ^{129}Xe and ^{131}Xe exhibit hyperfine

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structure since they have nonzero nuclear spin (^{129}Xe: $I = 1/2$ and ^{131}Xe: $I = 3/2$). In the optical spectrum, we can resolve the hyperfine splitting of both the metastable state and the optically excited state. The transmission spectrum of the sample at 823 nm shows four resonances of ^{129}Xe. The absorption by the even isotopes converge to the same peak. A small resonance from ^{131}Xe is visible in this spectrum as well. The transmission spectrum of the sample at 882 nm gives three resonances by ^{129}Xe, a resonance for even isotopes and a small resonance of ^{131}Xe. The hyperfine structure coefficient could be calculated from the distances between the resonances. We measure $A_{\text{metastable}} = -2.3844$ GHz, $A_{J=2} = -889.6$ MHz, $A_{J=3} = -873.2$ MHz, in agreement with the reported values [2, 3, 4].

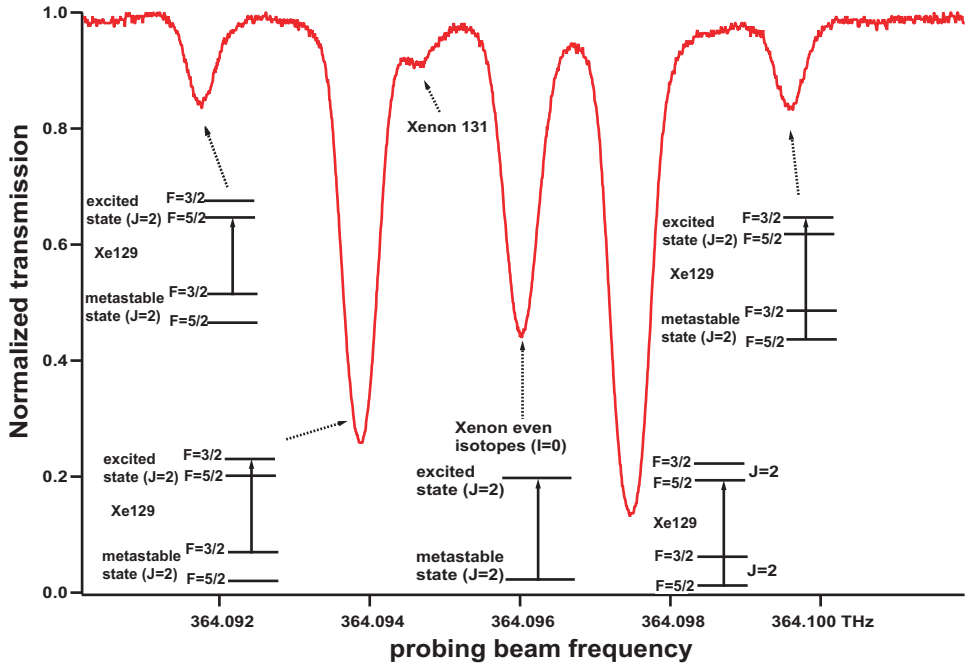

Figure 1: Transmission spectrum of metastable xenon at 823 nm optical excitaction.

2 Apparatus and experiments

Xenon gas is sealed in a pyrex cell (see Fig. 3). The part of the cell that is optically probed is cylindrically shaped, 4 inches long and 2 inches in diameter. The cell has two arms, one of which is connected to a pressure gauge monitoring the total pressure of the gas within the cell. The other arm goes to a liquid nitrogen cryostat. The pressure of Xe in the cell is determined by the temperature of the cryostat. Heater wire wound on the bottom of the arm which is cooled by the thermal bath controls this temperature. The cylindrical part of cell is placed in the isocenter of a set of Helmholtz coils which generates a homogeneous magnetic field. The light going through the cell and absorbed by metastable ^{129}Xe then focused by a convex lens and collected by a photodiode. The hyperfine resonance is stimulated by a microwave source generated by a synthesizer which is internally modulated at 10 kHz. This signal, at ≈ 5.96125 GHz, is amplified and coupled into a horn antenna and then beamed

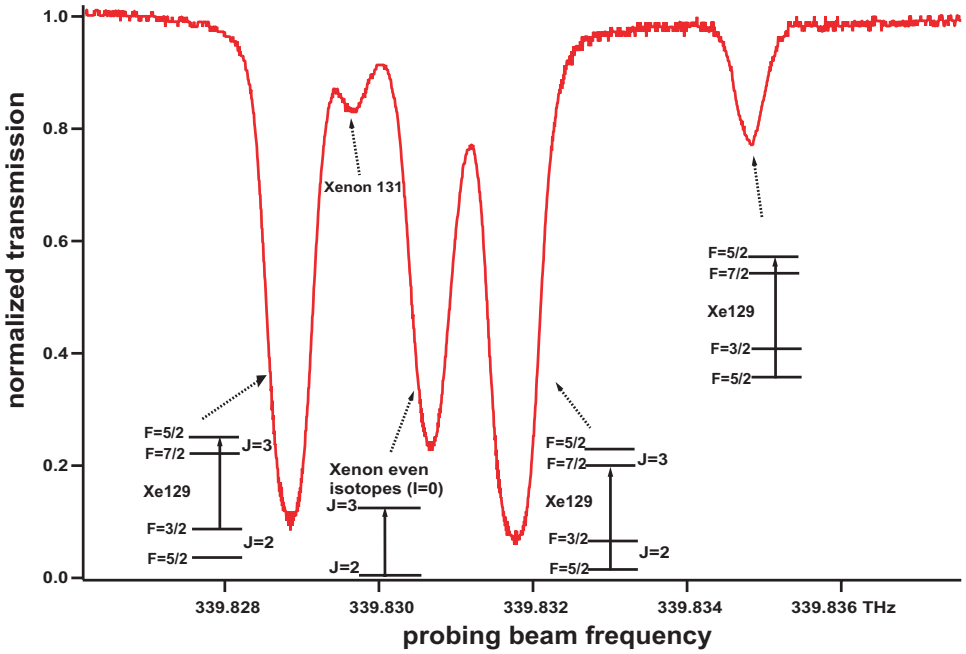

Figure 2: Transmission spectrum of metastable xenon at 882 nm optical excitaction.

on to the cell. The photodiode signal is amplified by a preamplifier operated in low pass mode and then by a lock-in amplifier which is locked by the modulation frequency of the microwave synthesizer. The output of the lock-in amplifier is monitored and recorded by a digital oscilloscope. When the Zeeman resonance signal is observed, the RF signal is applied by a solenoid. The solenoid is driven by a function generator running at ~ 5 MHz or ~ 3.3 MHz and internally modulated. The method to process the optical signal is the same as when we observe the hyperfine resonance signal.

3 Milestones

Our first attempts to measure Zeeman resonances revealed linewidths that were ≈ 500 kHz full width at half maximum (FWHM) in cells with Xe pressures of ≈ 1 torr, in the summer of 2007. By late 2007, we successfully acquired hyperfine resonance signals. A significant problem in our early work was the contamination of our cells with impurities, which affect the discharge properties of our cells. In early 2008 we identified hydrogen as a major source of contamination in our cells by using a residual gas analyzer (RGA, see Fig. 4). This allowed us to eliminate the effect of such contamination by adding a small hydrogen getter to our cells. The further problem of Xe being driven to and from the cell walls during discharge was mitigated by using a cryotrap design where the vapor pressure of Xe near 77 K (≈ 3 millitorr) was controlled by cryopumping the Xe. In our current design, we can routinely measure hyperfine linewidths of $\lesssim 50$ kHz FWHM.

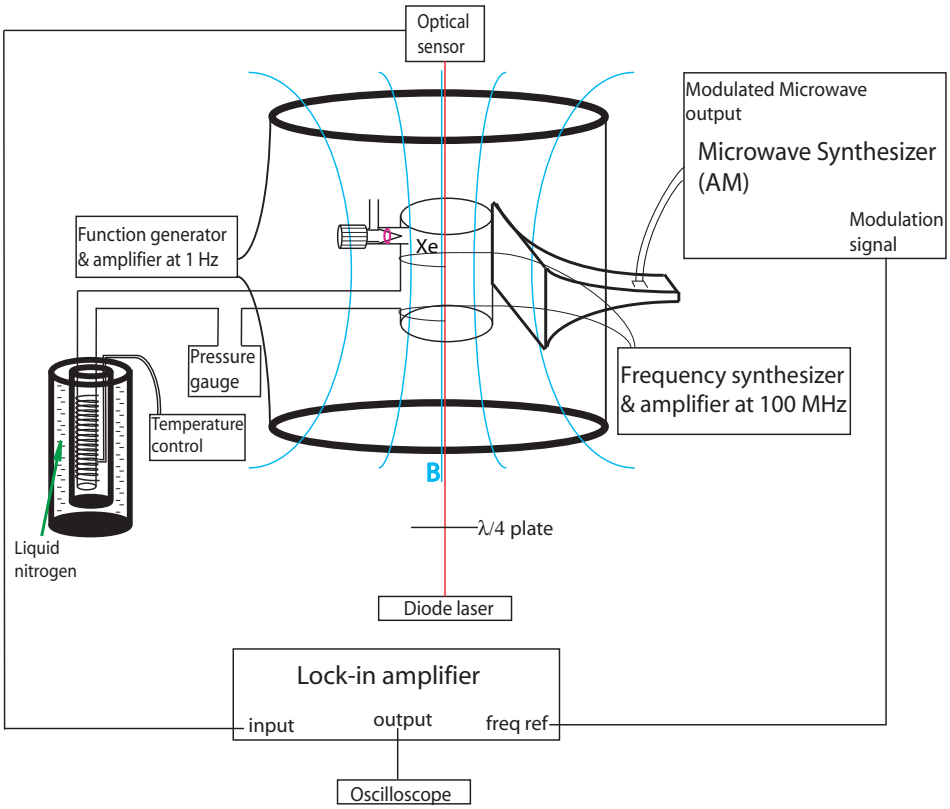

Figure 3: Diagram of the experimental apparatus.

4 Polarization of metastable ^{129}Xe

We measure the polarization of metastable ^{129}Xe by observing the absorption spectrum (see Fig. 5). The spectrum does not fit to a spin temperature distribution. We simulated the pumping, relaxation and microwave excitation processes to determine the population distribution and the corresponding spectrum. We compared the experimental result with series of simulated results to calculate the measured polarization.

In the simulation, we consider several relaxation mechanisms of the polarization including diffusion to the cell wall which completely destroys the atomic polarization, strong and weak J-damping which transfers electronic polarization to the angular momentum of the collisional pair but conserves the nuclear spin polarization, and metastability exchange in which the electronic polarization of the atoms is exchanged but the nuclei are unaffected. For the excited sublevels, we also considered relaxation processes that occur before the atoms repopulate the metastable state. In Fig. 6, the heights of the columns represent the relative populations of the quantum states when the atoms are just optically pumped but without microwave signal applied.

In this spectrum as shown in Fig. 5, we have eight resonances over the 35 MHz sweeping range of the microwave frequency around the hyperfine splitting while the longitudinal field is about 4.55 Gauss. The eight resonances correspond to eight permitted σ transitions between hyperfine multiplets $F = 3/2$ and $F = 5/2$. The longitudinal magnetic field B makes

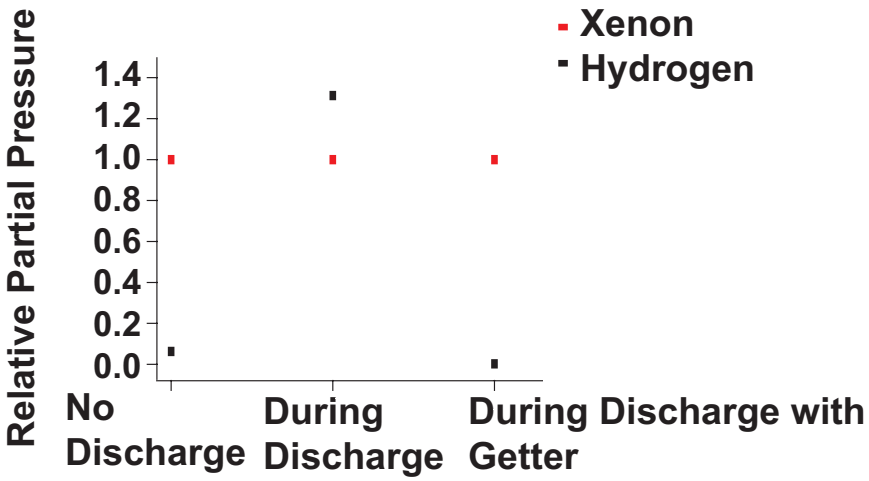

Figure 4: The relative partial pressures of molecular hydrogen (H_2) and ^{129}Xe measured on our RGA before and during discharge, and during discharge with a hydrogen getter inside the cell. To a lesser extent, we also measured water and nitrogen contaminants in the cells. Using the hydrogen getter eliminates these problems.

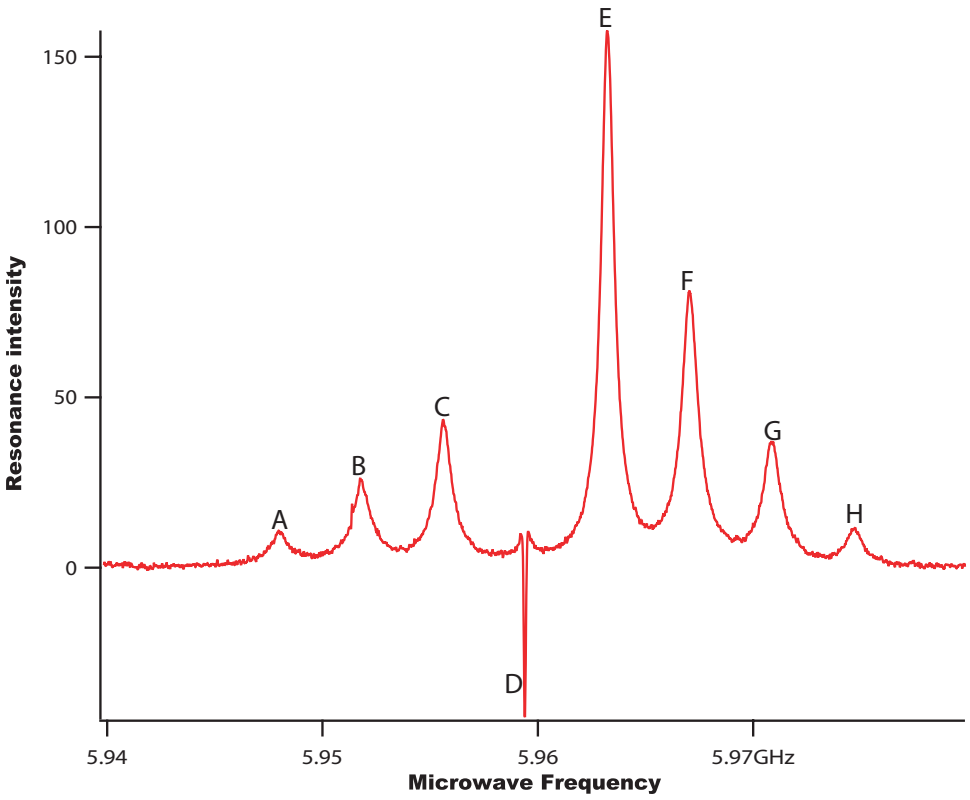

Figure 5: Hyperfine spectrum of metastable ^{129}Xe. The calculated polarization is 22.4%. The transitions A, B, etc. are shown in Fig. 6. The dip in line D is due to the change in population difference (see Fig. 6) between this resonance and the others. It is narrow because the $m = -\frac{5}{2}$ sublevel is dark, and the bumps above zero in this resonance are caused by the presence of both σ and π microwave transitions.

magnetic sublevels' Zeeman splitting gyromagnetic ratio $\frac{\nu}{B} = \frac{\mu_B g_F}{h}$. The gyromagnetic ratios of the two hyperfine multiplets are 2.5193 MHz/G and 1.6795 MHz/G for $F = 3/2$ and $F = 5/2$ respectively. Simulations with the same spectrum as the experimental data give a calculated polarization of the nuclear spin of the metastable ^{129}Xe as 22.49%.

5 Pressure Broadening of the Resonances

Measurements of the pressure broadening of the resonance lines are shown in Fig. 7. Also shown are linear fits of the linewidths vs. pressure. The values for these fits are given in Table 1. Many measurements were taken, and we believe the scattering is due to a combination of effects including varying levels of contamination and helium permeation into the cell. Nevertheless, for the end resonance cases pumping both of the strong optical transitions of Fig. 1, both the slope and the intercept are identical as is seen in Table 1 and Fig. 7.

Although the ratio of the slopes we measure for the Zeeman resonances is similar to those measured previously [5], the measured slopes are different. The previous experiments involved pure ^{129}Xe, and give, for example, a slope of 5.76 ± 0.14 kHz/millitorr for the

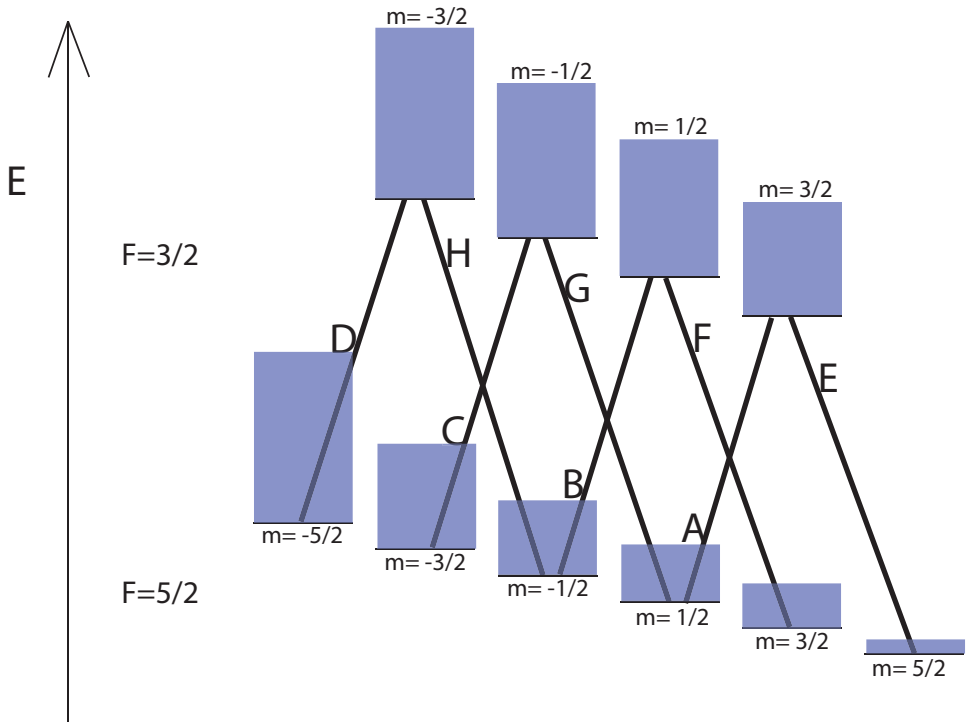

Figure 6: Illustration of metastable ^{129}Xe polarization.

$F = \frac{3}{2}$ Zeeman resonance while our value is 7.7 ± 0.3 kHz/millitorr. Part of this difference is presumably due to the increased slowing down of the relaxation that will happen when the relative abundance of ^{129}Xe is increased. In our experiments we use only 86% ^{129}Xe and the rest is nearly all even Xe isotopes, with only a small fraction being ^{131}Xe. Previous reports on Zeeman resonances in even isotopes of Xe have given slopes that vary by a factor of $2-3$.

Resonance	Slope	Intercept
$F = \frac{5}{2}$ Zeeman	5.9 ± 0.3	24.5 ± 1.9
$F = \frac{3}{2}$ Zeeman	7.7 ± 0.3	27.0 ± 2.0
End resonance	6.6 ± 0.1	22.8 ± 0.9
2nd resonance	6.4 ± 0.4	26.5 ± 2.1
3rd resonance	6.8 ± 0.6	26.8 ± 3.5
End resonance (pump $F = \frac{5}{2}$)	6.7 ± 0.1	22.3 ± 0.9

Table 1: Fit coefficients for the data in Fig. 7. In Fig. 6, the corresponding resonances are: end resonance (E), 2nd resonance (F), and 3rd resonance (G). The 4th resonance (H) is too weak to get useful linewidth information from.

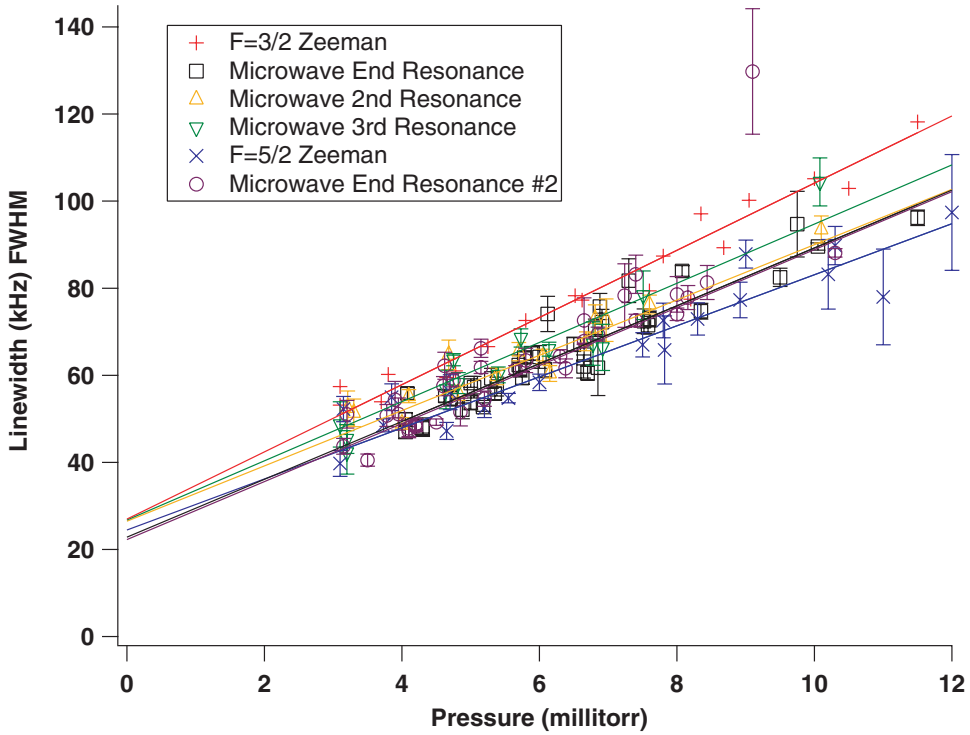

Figure 7: Measured linewidths (FWHM) for metastable ^{129}Xe hyperfine and Zeeman resonances, in kHz. The measured pressure is in millitorr. Microwave end resonance #2 refers to the data acquired while pumping the $F = \frac{5}{2} \rightarrow F = \frac{5}{2}$ optical transition (see Fig. 1). All other data except the $F = \frac{5}{2}$ Zeeman resonance correspond to pumping the $F = \frac{3}{2} \rightarrow F = \frac{3}{2}$ optical transition. The coefficients of the linear fits are given in Table 1.

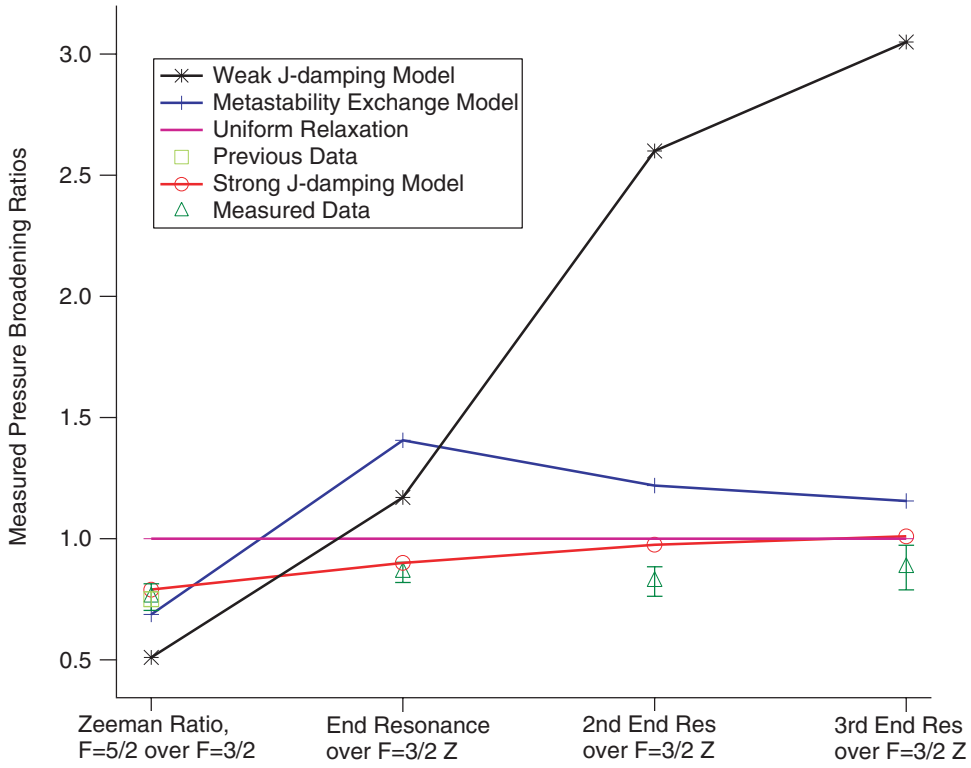

Figure 8: Measured and modeled pressure broadening ratios. The experimental data are the slope of the lines from Fig. 7 and Table 1. The previous data represents a measurement in pure ^{129}Xe from [5]. The modeling includes weak J-damping, strong J-damping, and metastability exchange, as described in Sect. 4. The end and second hyperfine resonances actually also include contributions from $\Delta m = 0$ transitions, which confounds the interpretation of the data somewhat.

6 Future Work

Future work with metastable ^{3}He could be pursued. ^{3}He has a different electronic structure than the other metastable noble gases, and its intrinsic relaxation is about an order of magnitude slower. Metastable ^{4}He is already used in portable magnetometry, and the hyperfine frequency of ^{3}He is almost the same as ^{87}Rb. A potentially more serious problem for a ^{3}He clock in comparison with one made with ^{129}Xe would be the increased effect of wall relaxation due to the more rapid motion of the helium atoms.

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